527 Rec'd PCT/PTC 06 NOV 2000

FORM PTO-1390 (REV. 5-93) U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

ATTORNEY'S DOCKET NUMBER 22750/459

TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371

U.S. APPLICATION NO. (If known, see 37 CFR 1.5) 09/674799

INTERNATIONAL FILING DATE PRIORITY DATE CLAIMED: INTERNATIONAL APPLICATION NO. (18.05.98)(31.03.99)PCT/EP99/02202 31 March 1999 18 May 1998 TITLE OF INVENTION **FUEL CELL** APPLICANT(S) FOR DO/EO/US BARTH, Thomas; SEVERICH, Birgit; and KASPER, Klaus Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information This is a FIRST submission of items concerning a filing under 35 U.S.C. 371. 1. This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371. 2. 🗆 This express request to begin national examination procedures (35 U.S.C. 371(f)) immediately rather than delay examination until the 3. ☒ expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1). A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date. 4. ⊠ A copy of the International Application as filed (35 U.S.C. 371(c)(2)) gh a. \square is transmitted herewith (required only if not transmitted by the International Bureau). b. ☑ has been transmitted by the International Bureau. 4 c. \square is not required, as the application was filed in the United States Receiving Office (RO/US) ũ 6. ⊠ A translation of the International Application into English (35 U.S.C. 371(c)(2)). **Z**. 🛛 Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)) a. \square are transmitted herewith (required only if not transmitted by the International Bureau). b. \square have been transmitted by the International Bureau. LF) c. \square have not been made; however, the time limit for making such amendments has NOT expired. have not been made and will not be made. A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)). An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)) (unsigned). \boxtimes A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)). 10. 🗆 Items 11. to 16. below concern other document(s) or information included: 11. An Information Disclosure Statement under 37 CFR 1.97 and 1.98. 12. An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included. 13. A FIRST preliminary amendment. A SECOND or SUBSEQUENT preliminary amendment. 14. 🗌 A substitute specification. 15. 🔲 A change of power of attorney and/or address letter. Other items or information: Translation of the International Search Report; Translation of the International Preliminary Examination 16. Report; first page of published International Application WO 99/60649; and two (2) sheets of drawings.

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| 17. ☑ The following fee | es are submitted: | | | CALCULATIONS | PTO USE ONLY |
| Basic National Fee (37 CFR 1.492(a)(1)-(5)): | | | | | |
| Search Report has b | peen prepared by the EP | O or JPO | \$860.00 | | |
| International prelimir | nary examination fee pai | d to USPTO (37 CFR 1.4 | 182) \$690.00 | | |
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| Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO | | | | | |
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| Surcharge of \$130.00 for furnishing the oath or declaration later than 20 30 months from the earliest claimed priority date (37 CFR 1.492(e)). | | | \$ | | |
| Claims | Number Filed | Number Extra | Rate | | |
| Total Claims | 18 - 20 = | 0 | X \$18.00 | \$ | |
| Independent Claims | 1 - 3 = | 0 | X \$80.00 | \$ | |
| Multiple dependent claim(s | s) (if applicable) | | + \$270.00 | \$ | |
| | TOTA | L OF ABOVE CAL | CULATIONS = | \$ 860.00 | |
| Reduction by ½ for filing by small entity, if applicable. Verified Small Entity statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28). | | | \$ | | |
| SUBTOTAL = | | | \$ 860.00 | | |
| Processing fee of \$130.00 for furnishing the English translation later the 20 30 months from the earliest claimed priority date (37 CFR 1.492(f)). | | | \$ | | |
| TOTAL NATIONAL FEE = | | | \$ 860.00 | | |
| Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be | | | | | |
| accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property + | | | \$ | | |
| TOTAL FEES ENCLOSED = | | | \$ 860.00 | | |
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| b. 🗵 Please charge my Deposit Account No. 11-0600 in the amount of \$860.00 to cover the above fees. A duplicate copy of this | | | | | |
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| Account No. 11-0600 . A duplicate copy of this sheet is enclosed. | | | | | |
| NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status. | | | | | |
| SEND ALL CORRESPONDENCE TO: SIGNATURE | | | | | |
| ; Kenyon & Kenyon | | | | | |
| One Broadway New York, New York 10004 Richard L. Mayer, Reg. No. 22,490 NAME | | | | | |
| 11/6/00 | | | | | |

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[22750/459]

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Inventors

Thomas BARTH et al.

Serial No.

To Be Assigned

Filed

Herewith

For

FUEL CELL

Examiner

To Be Assigned

Art Unit

To Be Assigned

Assistant Commissioner for Patents Washington, D.C. 20231

PRELIMINARY AMENDMENT

SIR:

Kindly amend the above-identified application, which is the national stage in the U.S. of PCT international application PCT/EP99/02202, before examination, as set forth below.

IN THE SPECIFICATION:

On page 1, line 1 delete "Description" and insert as a heading:

--BACKGROUND OF THE INVENTION--.

On page 1, line 3 delete "Technical area" and insert as a heading: --<u>FIELD OF THE INVENTION</u>--.

On page 1, line 14 delete "Background Information" and insert as a heading: --DESCRIPTION OF RELATED ART--.

On page 1, line 23 delete "Explanation of the Invention" and insert as a heading: --<u>SUMMARY OF THE INVENTION</u>--.

On page 2, before line 1 insert:

--BRIEF DESCRIPTION OF THE DRAWINGS

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Figure 1 is a perspective, cross-sectional view of an exemplary embodiment of a fuel cell in accordance with the invention.

Figure 2 is a graph of current against voltage curves corresponding to Examples 1, 3 and 5.

DETAILED DESCRIPTION OF THE INVENTION ---.

IN THE CLAIMS:

On page 12, the first line delete "Patent Claims" and insert as a heading: --WHAT IS CLAIMED IS:--.

Cancel original claims 1-20, without prejudice, and insert new claims 21-38 as follows:

-21. (New) A fuel cell, comprising: a housing, at least a first layer (1) made of a proton-conductive material, gas-permeable electrodes (2), and second layers (3) arranged on either side of the first layer (1) in the form of electrically conductive plates, which contact in an electrically conductive manner the electrodes (2) at closely adjacent intervals and which, together with the electrodes (2), define gas-carrying channels (4), the first layer (1) or second layers (3) having an essentially planar surface, the first layer (1) or the second layers (3) being provided with a corrugation, pleating, or debossing to form the channels (4), wherein: the first layer (1) is made of a polymer material and is covered on both sides by a catalyst layer, the electrodes are arranged on the catalyst layers, and the first layer (1) is made of a porous foil, a web, a weft, or a non-woven fabric made of short or endless fibers and is impregnated with an electrolyte to the point of saturating the pores.

22. (New) The fuel cell according to Claim 21, wherein the first layer (1), made of a porous foil, a web, a weft, or a non-woven fabric made of short or endless fibers, is chemically and physically inert in the presence of the electrolyte up to a temperature of 200°C.

- 23. (New) The fuel cell according to Claim 21, wherein the first layer is made of nonwoven fabric, wherein the nonwoven fabric is made of microfibers, film fibrilla, or microfilaments.
- 24. (New) The fuel cell according to Claim 21, wherein the first layer (1) is made of PTFE or polysulphone.
- 25. (New) The fuel cell according to Claim 21, wherein the first layer (1) has an average pore radius of 20 nm to $10 \mu m$.
- 26. (New) The fuel cell according to Claim 21, wherein the first layer (1) is filled with the perfluorinated ionomer.
- 27. (New) The fuel cell according to Claim 26, wherein the perfluorinated ionomer is a polytetrafluoroethylene having sulphonated perfluorovinylether side chains.
- 28. (New) The fuel cell according to Claim 21, wherein the microfiber non-woven fabric is impregnated with an aqueous sulfuric acid solution of from 1 to 5 moles.
- 29. (New) The fuel cell according to Claim 21, wherein the microfiber nonwoven fabric is saturated using concentrated phosphoric acid.
- 30. (New) The fuel cell according to Claim 21, wherein the microfiber nonwoven fabric is saturated using hydrated zirconium phosphate and ammonium dihydrogen phosphate.
- 31. (New) The fuel cell according to Claim 22 , wherein the first layer (1) has a mass per unit area of 20 to 200 g/m², a thickness, when impregnated by the electrolyte, of \leq 1 mm, an average pore radius of 0.1 to 10 μ m, and a pore volume of from 65 to 92%.
- 32. (New) The fuel cell according to Claim 21, wherein the first layer (1) is laminated together with the electrodes (2).

- 33. (New) The fuel cell according to Claim 32, wherein the electrodes (2) and the first layer (1) are bonded to each other in surface areas having a distance from each other.
- 34. (New) The fuel cell according to Claim 21, wherein the electrodes (2) are made of carbonized fibers of polymer material.
- 35. (New) The fuel cell according to Claim 32, wherein the electrodes (2), at a thickness of less than 0.5 mm, have a mass to area ratio of 20 to 100 g/m^2 .
- 36. (New) The fuel cell according to Claim 21, wherein the second layer (3) is made of sheet metal.
- 37. (New) The fuel cell according to Claim 21, wherein the channels (4) have through-flow in accordance with the countercurrent principle.
- 38. (New) The fuel cell according to Claim 21, wherein the channels (4), at at least one end, terminate in through-bores from projections of the housing that mesh with each other, cog-like, sealingly contacting on both sides the layer (1, 3) that is provided with the corrugation, pleating, or debossing.--

IN THE ABSTRACT:

On page 15, line 1 delete "Abstract" and insert as a heading: --ABSTRACT OF THE DISCLOSURE--.

Remarks

This Preliminary Amendment cancels, without prejudice, original claims 1-20 of the underlying PCT Application No. PCT/EP99/02202, and adds new claims 21-38. The new claims conform the claims to U.S. Patent and Trademark Office rules and do not add new matter to the application.

The above amendments to the specification and abstract are to conform the specification and abstract to U.S. Patent and Trademark Office rules or to correct informalities, and do not introduce new matter into the application.

The underlying PCT Application No. PCT/EP 99/02202 includes an International Search Report, dated September 9, 1999. The Search Report includes a list of documents that were uncovered in the underlying PCT Application. A copy of the Search Report is included herewith.

The underlying PCT Application also includes an International Preliminary Examination Report, a copy of which is included herewith.

Applicants respectfully submit that the subject matter of the present application is new, non-obvious, and useful, as indicated in the Preliminary Examination Report. Prompt consideration and allowance of the application are respectfully requested.

Respectfully submitted,

Dated: 11/6/65

Richard L. Mayer Reg. No. 22,490

> KENYON & KENYON One Broadway New York, NY 10004 (212) 425-7200

FUEL CELL

Description

Technical area

The invention relates to a fuel cell, having at least a housing, at least one layer made of a proton-conductive, polymer material, the layer being covered on both sides by catalyst layers, gas-permeable electrodes on the catalyst layers, and bipolar plates which contact the electrodes in an electrically conductive manner at closely adjoining intervals and which, together with the electrodes, define gas-conveying channels.

Background Information

A fuel cell of this type is known from [the publication] Spectrum of Science, July 1995, p. 98. The channels, in this context, are molded into the bipolar plates and run parallel to each other. Their manufacture is accordingly cumbersome and expensive. In addition, the power-weight ratio of the known fuel cells is unsatisfactory.

Explanation of the Invention

- The present invention is based on the objective of making available a fuel cell of the type cited above that can be manufactured in a manner that is technically simpler and more cost-effective.
- This objective is achieved on the basis of the present invention in a fuel cell of the type mentioned above having the characterizing features of Claim 1. The subclaims refer to advantageous embodiments.

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In the fuel cell according to the present invention, it is provided that the first layer or the second layers for creating the channels is/are provided with a corrugation, a pleating, and/or a debossing and that the respective other layer or layers is/are configured in a planar manner. The surfaces of the first and the second layers, situated opposite each other, run parallel to each other with the result that there is either an enlargement of the active surface or a reciprocal overlapping of the profiles of laterally adjacent channels, resulting in a significant increase in the power density per space unit and therefore also in an improvement in the power-weight ratio. This is a great advantage particularly in applications in the mobile area.

Which of the two layers is provided with the corrugation, pleating, and/or debossing, and which is given a planar configuration, has no further significance in the meaning of the present invention. However, for technical reasons, a design is preferred in which the first layer, including the catalyst layers and electrodes arranged thereon, is configured in a planar manner and the second layer, made mostly of metal, has the corrugation, pleating, and/or debossing. The functional reliability of designs of this type is generally greater than in the case of other variants.

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The cross-section of the fuel cell can be configured similarly to corrugated paper, in which a corrugated, pleated, or debossed layer alternates with a planar layer, until the desired overall thickness or overall strength is reached. The successive channels between the individual layers, during normal use, have flowing through them alternately a gas containing hydrogen and one containing oxygen, the gas at the same time being able to act to remove from the fuel cell excess heat and the water arising during the chemical reaction. For this purpose, it is advantageous if at least those channels in which water builds up are arranged in a vertical manner during normal use and have a gas flowing

through them from the top to bottom. If the temperature of the fuel cell is sufficiently high, the water exists in the form of gas, i.e., in the form of water vapor. In such a case, the actual water separation can be performed outside of the channels, and the remaining unused gas, if appropriate, can be tempered and returned to the channels in a circulation circuit. The remaining channels advantageously run parallel to the "water-carrying" channels. However, they can also run transverse to these channels.

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In embodiments in which either the first or the second layer is elastically compressible and the other layer, managing in each case to mesh with the former, is configured so as to be incompressible, the result is automatically a good seal between the adjoining channels, which makes it possible to compensate for inaccuracies in the manufacturing process and to achieve a high degree of efficiency. If the corrugation, pleating, and/or debossing constitute a component of the compressible layer, then production is especially simple. It can take place using methods that are applied in textile processing, for example using deep drawing or pleating methods. In sheet metal processing, similar methods are known. They can be applied given analogous conditions.

The first layer can be composed of a porous foil, a web, a weft [knitted fabric], or a non-woven fabric made up of short or endless fibers, and the first layer is filled to saturation with perfluorinated ionomer, the perfluorinated ionomer potentially being a polytetrafluoroethylene having sulphonated perfluorovinylether side chains. Alternatively, it is possible to fill the microfiber non-woven fabric using an aqueous sulfuric acid solution of from 1 to 5 moles, or using concentrated phosphoric acid. Furthermore, it is possible to use hydrated zirconium phosphate and ammonium dihydrogen phosphate.

As the first layer becomes thinner, the efficiency of the fuel

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cell can be improved. From this point of view, it has proven to be advantageous if a non-woven fabric contained in the first layer is made of microfibers, film fibrilla, or microfilaments. However, it is also possible to use porous foils. As materials, PTFE and polysulphone have proven to be especially effective.

When a microfiber non-woven fabric is used as the proton conductor, it is provided that the latter is impregnated using an electrolyte to the point of saturation, the microfiber non-woven fabric being chemically and physically inert with respect to the electrolyte at temperatures up to $+200^{\circ}$ C as well as under oxidizing and reducing conditions, the weight of the microfiber non-woven fabric amounting to 20 to 200 g/m²; the thickness of the non-woven fabric amounting to a maximum of 1 mm and the pore volume amounting to 65 to 92%.

The average pore radius of the microfiber non-woven fabric should be from 20 nm to 10 μm .

The first layer can be laminated together directly with the electrodes, for example in a direct, reciprocal adhesion in surface areas having a distance from each other. In this context, the electrodes can be formed by carbonized fibers made of polymer material, for example, carbonized polyacrylonitrile or pitch fibers, and, assuming a thickness of less than 0.5 mm, they can have a mass per unit area of 20 to 100 g/m^2 .

In the simplest case, the second layer is made of a metallic sheet of planar configuration. The heat arising in the fuel cell during normal use can be removed via this [second layer] parallel to the generated current, either by itself or as a supplement.

The channels, irrespective of the specific configuration of the first and second layer, can emerge, at least at one end,

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in through-bores from projections of the housing that mesh with each other, cog-like, sealingly contacting on both sides the layer provided with the corrugation, pleating, and/or debossing. As a result, it is assured that the reaction gases that are conveyed through the fuel cell during normal use can travel through the first layer to the reaction point only as a result of proton conduction.

In Figure 1, an exemplary embodiment of the fuel cell is represented. It includes an undepicted housing, a first layer 1 made of a proton-conductive, polymer material, that is covered on both sides by catalyst layers, gas-permeable electrodes 2 on the catalyst layers and arranged on both sides of first layer 1, second layers 3 in the form of electrically conductive plates, which contact in an electrically conductive manner electrodes 2 at closely adjacent intervals and which, together with electrodes 2, define the boundary of gascarrying channels 4, first layer 1 being provided with a pleating for the formation of channels 4 and second layers 3 being formed by planar sheets of metal that contact the folded edges of electrodes 2 abutting against first layer 1. During normal use, gases containing oxygen and hydrogen are conveyed through channels separated by first layer 1, and, after reaction on the catalyst surface, are brought, through first layer 1, to react with each other. The current arising in this process is conveyed via electrodes 2 to second layers 3 and via them is carried off. The heat released during the chemical process follows the same path.

The fuel cell depicted schematically in the drawing shows the smallest functional unit. Its current production is a function of the size of the first layer as well as of the cross-section of channels 4. Energy production can be increased by increasing the corresponding values or by connecting in parallel in one self-enclosed packet units that are shaped in accord with each other.

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In the simplest case, it is provided in the fuel cell according to the present invention that the first layer is furnished with a corrugation, pleating, and/or debossing for the formation of the channels and that second layers 3 are configured in planar fashion and are formed by metal sheets. In this context, it is advantageous if first layer 1 contains a non-woven fabric made of short or endless fibers to give first layer 1 the necessary mechanical capacity for resistance. As a result, the electrolyte contained in the hollow spaces of a non-woven fabric of this type is not subjected to mechanical stress. The electrolyte can be optimized with respect to type and amount, in order to achieve especially high electrochemical effectiveness. The use of a porous foil, whose pores contain an appropriate electrolyte, is also possible.

The temperature resistance of first layer 1 is determined as a function of the type of non-woven fabric, or porous foil, contained in it. A particularly high temperature resistance is achieved if in the first layer a non-woven fabric, or a porous foil made of PTFE or polysulphone, is used. The operating temperatures can exceed 90°C in one embodiment of this type without resulting in the production of catalyst poisons, in operation using reformed methanol, in the course of secondary chemical reactions, and without impairing the service life of the fuel cell. Non-woven fabrics made of microfibers have the advantage that the pore structure takes in an extremely large, proportional volume, that it is configured so that the pores merge into each other, and that large portions of the pores are covered in the direction of the surfaces of the fibers. In this manner, the undesirable leaching of the electrolyte by the water formed during normal use is suppressed.

The layer made of polymer material can be formed by a microfiber non-woven fabric impregnated with an electrolyte to the point of saturation or by a plastic foil that is sintered together or stretched, generating the pores. The fibers, or

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the foil, in this context, are made of a polymer material that is chemically inert in the presence of the electrolyte under conditions of normal use, temperatures up to +200°C being possible both under oxidizing as well as under reducing conditions. Particularly suitable for generating the layer is polytetrafluoroethylene.

The fibers can be configured so as to be endless and can be bonded without the use of secondary adhesives, for example using solvent welding and/or a fusion of the fibers merging into one another.

It is preferable to use a non-woven fabric having a tensile strength longitudinal/transverse of more than 50 MPa, an elongation at fracture of 50 to 100%, and an elastic modulus of 2 to 4 GPa which is physically resistant at ambient temperatures up to + 200°C. The non-woven fabric weight should be 20 to 200 g/m² at a thickness, when impregnated with the electrolyte, of less than \leq 1 mm, [and the fabric should have] an average pore radius of 0.1 to 10 μ m and a pore volume of 65 to 92%. The dielectric constant can be 0.3200 to 3500 Hz.

The non-woven fabric framework assures the mechanical strength of the membrane so that the electrolyte is no longer called on to satisfy this objective and therefore need be used only in significantly smaller concentrations, for controlling the electrochemical processes in the cell. As a result, the material costs for the membrane are reduced by up to 90%, compared, e.g., with the expense of manufacturing a correspondingly dimensioned foil made of perfluorinated ionomer.

The temperature resistance of the membrane according to the present invention, if not governed by other factors, is essentially determined by the nonwoven fabric material. This circumstance makes it possible to employ the membrane also in fuel cells that are operated using reformed methanol; in

addition, the quantity of catalyst poisons arising in the course of the chemical secondary reactions is in any case reduced at operating temperatures over 90°C, which results in a greater service life for the cell.

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The following examples illustrate that the present invention in different variants is always superior to a pure polymer membrane made of perfluorinated ionomer. The base materials described below are common to all of the examples:

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Nonwoven fabric material: polysulphone fibers having a rectangular cross-section (width 6 to 13 $\mu \rm m$, height 1.7 to 2.4 $\mu \rm m)$.

Mechanical characteristics of the polysulphone material:

melting range: 343 to 399°C.

Tensile strength: 70 MPa

Elongation to fracture: 50 to 100%

Elastic modulus: 2.4 GPa

Bending temperature under 1.8 MPa load: 174°C

Dielectric constant: 3100 Hz

Manufacturing the fibers: spinning a solution of polysulphone in methylene chloride in the electrostatic field. For example, an apparatus according to German Laid-Open Print 26 20 399 can be used for this purpose. The fibers are gathered on a textile holder that is in continual linear motion.

Nonwoven fabric properties:

Weight: 150 g/m²

30 Thickness (compressed): 0.05 mm

Thickness (impregnated with electrolyte): 0.25 mm

Average pore radius in uncompressed state: 8 $\mu \mathrm{m}$

Average pore radius in compressed state: 4 μm

Pore volume: 83%

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The temperature resistance of the membrane according to the present invention, if not governed by other factors, is

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essentially determined by the nonwoven fabric material and, for the pure fiber material polysulphone, accordingly ends only at roughly 174°C. As a consequence of the mechanical bonding of the fibers to each other in the nonwoven fabric, the mechanical stability increases, additionally up to temperatures of 250°C. In this way, a high-temperature operation of the fuel cell is possible, which is significant, for example, for limiting the production of catalyst poisons.

Example 1

The micro-fiber nonwoven fabric is coated with liquid nafion, a commercial, perfluorinated ionomer made by the DuPont Co., in a glass frit having a 16 mm diameter. Through the application of a light underpressure [partial vacuum], the liquid phase is sucked into the pore structure of the nonwoven fabric. For removing solvents, the membrane impregnated in this manner is treated at 60°C in a drying chamber. Storage until further processing is possible in distilled water.

Examples 2 through 4:

The microfiber non-woven fabric is impregnated with three aqueous sulfuric acid solutions, of different moles, by analogy to Example 1, the sulfuric acid, however, being heated to roughly 70°C for reducing the viscosity. Without obtaining a different result, the nonwoven fabric can also be boiled for a few minutes in the acid heated to 70°C.

The membrane obtained in this manner is stored advantageously in the corresponding impregnating medium.

For the membranes prepared in this manner, the following specific conductivities were determined using a method in accordance with DIN 53779, dated March 1979:

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| Example | Measuring Temp °C | spec. conductivity S/cm |
|------------------------------------|-------------------|-------------------------|
| 1 | 23 | 0,016 |
| 2 | | |
| 1 M H ₂ SO ₄ | 18 | 0.031 |
| 3 | | |
| 3 M H ₂ SO ₄ | 18 | 0.041 |
| 4 | | |
| 5 M H ₂ SO ₄ | 18 | 0.080 |
| 5 | | |
| (comparative | | |
| basis) | 25 | 0.070 |

Example 5 in the table represents a comparative example for corresponding measurements on a 125 μm thick, self-supporting polymer membrane of the related art, made of perfluorinated ionomer (nafion-117, DuPont).

The values for the specific conductivity S/cm clearly show that, using the mechanically more resistant membrane according to the present invention, which is significantly more costeffective than pure nation and is simpler in design, it is possible to operate a fuel cell at power levels corresponding to the related art.

In comparison to a swollen nafion membrane of, e.g., 125 $\mu\rm m$ thickness, the nonwoven fabrics impregnated with electrolyte and used in Examples 1 through 4 are twice as thick.

The power of the fuel cell, represented by the product of voltage and amperage, can be reached not only using higher acid concentrations, i.e., higher specific conductivities S/cm, but also as a result of the diffusion inhibition from using thinner non-woven fabrics.

For example, in Figure 2, the corresponding current/voltage

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curves are illustrated at room temperature, which correspond to Examples 1, 3, and 5. The result is that, compared with the related art (Example 5), comparable curve shapes are achieved using the membranes according to the present invention. The effect of a higher cell output, mentioned above, from higher acid concentrations, or thinner nonwoven fabric materials, would result, in this illustration, in a shift of the curves in the positive direction of the ordinates.

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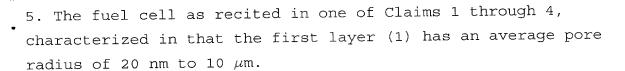
Patent Claims

1. A fuel cell, including a housing, at least a first layer (1) made of a proton-conductive material, gas-permeable electrodes (2), and second layers (3) that are arranged on either side of the first layers (1) in the form of electrically conductive plates, which contact in an electrically conductive manner the electrodes (2) at closely adjacent intervals and which, together with the electrodes (2), define gas-carrying channels (4), the first layer (1) or second layer (3) having an essentially planar surface, the first layer (1) or the second layers (3) being provided with a corrugation, pleating, and/or debossing, to form the channels (4),

characterized in that the first layer (1) is made of a polymer material and is covered on both sides by catalyst layers, that the electrodes are arranged on the catalyst layers and that the first layer (1) is made of a porous foil, a web, a weft, or a non-woven fabric made of short or endless fibers and is impregnated using an electrolyte to the point of saturating the pores.

- 2. The fuel cell as recited in Claim 1, characterized in that the first layer (1), made of a porous foil, a web, a weft, or a non-woven fabric made of short or endless fibers, is chemically and physically inert in the presence of the electrolyte up to a temperature of 200°C.
- 3. The fuel cell as recited in Claim 1 or 2, in which the first layer is made of nonwoven fabric, characterized in that the nonwoven fabric is made of microfibers, film fibrilla, or microfilaments.
- 4. The fuel cell as recited in one of Claims 1 through 3, characterized in that the first layer (1) is made of PTFE or polysulphone.

REVISED PAGE



- 6. The fuel cell as recited in one of Claims 1 through 5, characterized in that the first layer (1) is filled with the perfluorinated ionomer.
- 7. The fuel cell as recited in Claim 6, characterized in that the perfluorinated ionomer is a polytetrafluoroethylene having sulphonated perfluorovinylether side chains.
- 8. The fuel cell as recited in one of Claims 1 through 7, characterized in that the microfiber non-woven fabric is impregnated with an aqueous sulfuric acid solution of from 1 to 5 moles.
- 9. The fuel cell as recited in one of Claims 1 through 7, characterized in that the microfiber nonwoven fabric is saturated using concentrated phosphoric acid.
- 10. The fuel cell as recited in one of Claims 1 through 7, characterized in that the microfiber nonwoven fabric is saturated using hydrated zirconium phosphate and ammonium dihydrogen phosphate.
- 11. The fuel cell as recited in one of Claims 2 through 10, characterized in that the first layer (1) has a mass per unit area of 20 to 200 g/m², a thickness, when impregnated by the electrolyte, of \leq 1 mm, an average pore radius of 0.1 to 10 μ m, and a pore volume of from 65 to 92%.
- 12. The fuel cell as recited in one of Claims 1 through 11, characterized in that the first layer (1) is laminated together with the electrodes (2).

- 13. The fuel cell as recited in Claim 12, characterized in that the electrodes (2) and the first layer (1) are bonded to each other in surface areas having a distance [from each other].²
 - 14. The fuel cell as recited in one of Claims 1 through 13, characterized in that the electrodes (2) are made of carbonized fibers of polymer material.
 - 15. The fuel cell as recited in one of Claims 12 through 14, characterized in that the electrodes (2), at a thickness of less than 0.5 mm, have a mass to area ratio of 20 to 100 g/m^2 .
 - 16. The fuel cell as recited in one of Claims 1 through 15, characterized in that the second layer (3) is made of sheet metal.
 - 17. The fuel cell has recited in one of Claims 1 through 16, characterized in that the channels (4) have through-flow in accordance with the countercurrent principle.
 - 18. The fuel cell as recited in one of Claims 1 through 17, characterized in that the channels (4), at at least one end, terminate in through-bores from projections of the housing that mesh with each other, cog-like, sealingly contacting on both sides the layer (1, 3) that is provided with the corrugation, pleating, and/or debossing.

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²Translator's note: The final phrase of this claim is translated literally. However, the meaning is somewhat obscure. The specification speaks of the close intervals/distances at which the first layer, having the electrodes bonded to it, sealingly contacts the planar side walls. Since the electrodes are bonded/laminated to the first layer, the distance could hardly refer to any existing between these two components.

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Abstract

A fuel cell, including a housing, at least one layer (1) made of a proton-conductive, polymer material, the layer being covered on both sides by catalyst layers, gas-permeable electrodes (2) arranged on the catalyst layers and on both sides of the first layer (1), second layers (3) in the form of electrically conductive plates, which contact in an electrically conductive manner the electrodes (2) at closely adjacent intervals and which, together with the electrodes (2), define gas-carrying channels (4), the one layer (1,3) contacting the other layer (3, 1) having an essentially planar surface, the first layer (1) or the second layers (3) for forming the channels (4) being provided with a corrugation, pleating, and/or debossing.

Fig.1

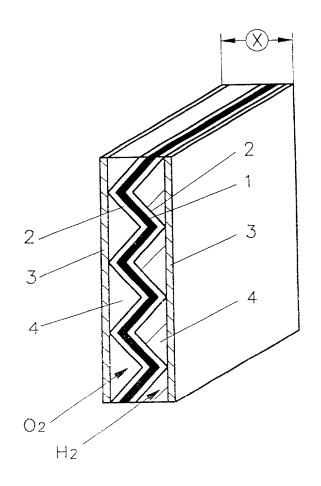
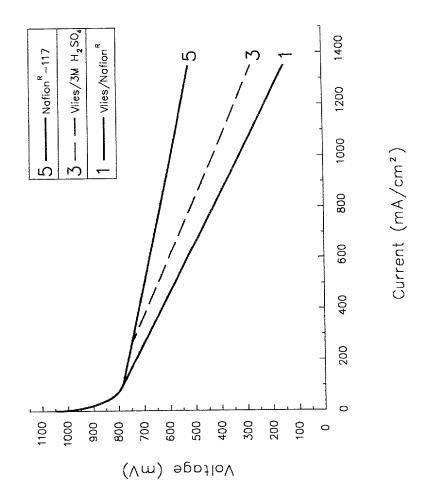


Fig.2



U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

DECLARATION AND POWER OF ATTORNEY

ATTORNEY'S DOCKET NO. **22750/459**

As a below named inventor, I hereby declare that:

My residence, post office address, and citizenship are as stated below next to my name,

I believe I am an original, first, and joint inventor of the subject matter that is claimed and for which a patent is sought on the invention entitled **FUEL CELL**, the specification of which was filed as International Application PCT/EP99/02202 on March 31, 1999.

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims.

I acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations, § 1.56(a).

PRIOR FOREIGN APPLICATION(S)

I hereby claim foreign priority benefits under Title 35, United States Code, § 119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

| COUNTRY | APPLICATION NUMBER | DATE OF FILING (day, month, year) | DATE OF ISSUE (day, month, year) | PRIORITY CLAIMED UNDER 35 U.S.C. § 119 |
|--------------------------------|-----------------------|--------------------------------------|-------------------------------------|--|
| Federal Republic of Germany | 198 21 985.7 | 18 May 1998 | | YES |

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorneys:

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I declare that all statements made herein of my own knowledge are true and all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under § 1001 of Title 18 of the United States Code and that such willful statements may jeopardize the validity of the application or any patent issuing thereon.

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